## Third Harmonic Generation of a Short Pulse Laser in a Tunnel Ionizing Plasma: Effect of Self-Defocusing

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**Abstract** Third harmonic generation of a Gaussian short pulse laser in a tunnel ionizing plasma is investigated. A Gaussian short pulse laser propagating through a tunnel ionizing plasma generates third harmonic wave. Inhomogeneity of the electric field along the wavefront of the fundamental laser pulse causes more ionization along the axis of propagation while less ionization off axis, leading to strong density gradient with its maximum on the axis of propagation. The medium acts like a diverging lens and pulse defocuses strongly. The normalized third harmonic amplitude varies periodically with the distance with successive maxima acquiring lower value. The self-defocusing of the fundamental laser pulse decays the intensity of the third harmonic pulse.

Keywords: third harmonic generation, self defocusing, laser, plasma.

### 1. INTRODUCTION

Harmonic generation of electromagnetic radiation in plasma has been a subject of extensive study for quite sometime [1-5]. The highly nonlinear nature of the interaction of the laser pulse with plasmas implies that harmonic light should be a significant feature of such interaction. Third harmonic generation is a very useful technique that can convert output if infrared lasers to shorter wavelength in the visible and near ultraviolet. Siders et al.[4] have demonstrated the generation of a tunable blue-shifted third harmonic generated at an ultra fast ionization front with efficiencies and spectral characteristic consistent with Vanin et al. [5]. Pressure tuning of the power spectrum centroid over the range  $10^{-5} < \Delta\omega / \omega \le 10^{-1}$ , corresponding to the pressure range ( $10 \le p \le 500 Torr$ ). In a nonlinear medium, an intense beam of finite diameter, which converge to a focus and then diverges, may exhibit nonlinear dynamics that significantly affect propagation dynamics. The greater the intensity, the bigger the difference between incoming and outgoing beams. Third harmonic generation with such an input beam produces third harmonic in the far field when the phase mismatch between the fundamental and its third harmonic is zero or negative (and small) [6]. They have also demonstrated this quantitatively for several cases.

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Rax and Fisch [7] have investigated relativistic third harmonic generation in a plasma. The phase velocity mismatch results in a third harmonic amplitude saturation and oscillation. In order to overcome this saturation, they described a phase matching scheme based on a resonant density modulation. Fedotov *et al.* [8] have observed third harmonic generation f unamplified 30-fs pulses of Cr: forsterite laser in a holey fiber.

They have demonstrated that high degree of light localization in the core of a holey fiber enhances third order nonlinear processes, allowing the third harmonic of non amplified 30 fs 0.2 nJ Cr: forsterite laser pulses appeared spectrally broadened at the output of the fiber. Lamprecht *et al.* [9] have shown that measuring the THG-ACF is a suitable method for evaluating the time characteristics of fs driven plasmon oscillations in metal nanoparticles. Resonant and off-resonant excitation of particle plasmons can be clearly distinguished by the ACF measurements.

Ganeev *et al.* [10] have studied harmonic generation from solid surfaces irradiated by 27 ps Nd:glass laser pulses in the intensity range  $10^{13}$ – $10^{15}$  W/cm². harmonic emission up to fourth order is observed in the specular reflection direction with conversion efficiencies of  $2 \times 10^{-8}$ , 10-10 and  $5 \times 10^{-12}$  for second, third, and fourth harmonic, respectively, for a p-polarized pump beam at  $10^{15}$  W/cm². Sodha *et al.* [11] have studied third harmonic generation in a collisional plasma by a Gaussian electromagnetic beam.

It is seen that the self-focusing of the main beam enhances the power of harmonic output by about three orders of magnitude in a typical case in the paraxial-ray approximation.

In this paper, we study the third harmonic generation of a Gaussian short pulse laser in a tunnel ionizing plasma. An intense short pulse laser with a Gaussian radial profile, propagating through a tunnel ionizing plasma. The plasma has radial as well as axial inhomogeneity. The ionization starts on the laser axis and creates a strong electron density gradient with its maximum on the axis of propagation. The medium acts like a diverging lens and pulse defocuses strongly. Laser imparts an oscillatory velocity to electrons and exerts a ponderomotive force on them, produces third harmonic nonlinear current density, driving third harmonic generation. The normalized third harmonic amplitude varies periodically with the distance with successive maxima acquiring lower value. The self-defocusing of the fundamental laser pulse decays the intensity of the third harmonic pulse.

In section II, we obtain an expression for the third harmonic transverse current density, deduce an equation for the third harmonic amplitude and solve this equation. A discussion of results is given in section III.

# 2. SELF-DEFOCUSING OF THE MAIN PULSE AND NONLINEAR THIRD HARMONIC CURRENT DENSITY

Consider the propagation of intense Gaussian laser short pulse in a tunnel ionizing plasma along z direction.

$$\vec{E}_1 = \hat{x}A_1(z,t)\exp[-i(\omega_1 t - k_1 z)] \qquad , \tag{1}$$

$$A_{1}^{2} = \frac{A_{10}^{2}}{f_{1}^{2}(z)} \exp[-r^{2}/r_{0}^{2}f_{1}^{2}] \quad , \tag{1a}$$

$$\vec{B}_{1} = \frac{c\vec{k}_{1} \times \vec{E}_{1}}{\omega_{1}} \quad , \tag{1b}$$

where  $r_0$  is the spot size of the main laser pulse at z=0, and  $f_1$  is the beam width parameter of the main laser pulse. The laser pump and the third harmonic electromagnetic wave obey the linear dispersion relation,  $k^2 \approx (\omega^2/c^2)(1-\omega_p^2/\omega^2)$ , where  $\omega_p = (4\pi n_0 e^2/m)^{1/2}$  is the plasma frequency, c is the velocity of light in vacuum,  $n_0$  is the electron density and -e and m are the charge and mass of an electron respectively.

The evolution of plasma frequency  $\omega p$  varies with time as<sup>12</sup>

$$\frac{\partial \omega_p^2}{\partial t} = \gamma [\omega_{pm}^2 - \omega_p^2] \quad , \tag{2}$$

where  $\omega_{pm}^2(z)=4\pi n_m e^2/m$ ,  $n_m$  is the initial density of neutral atoms,  $\gamma=(\pi/2)^{1/2}(I_0/\hbar)(|E|/E_A)^2\exp(-E_A/|E|)$ ,  $E_A=(4/3)\sqrt{2}mI_0^{3/2}$ , where  $I_0$  is the ionization potential and  $h=2\pi\hbar$  is the plank's constant. Following the Liu and Tripathi<sup>13</sup>, for paraxial ray approximation, we introduce dimension less quantities  $\xi=z'/R_d$ ,  $\eta=\gamma_{00}t'$ , where  $\gamma_{00}=(I_0/\hbar)(\pi/2)^{1/2}(1/g^2)$ , express  $A_1=A_{10}\exp(ikS)$  and expand relevant quantities, in the paraxial ray approximation, as  $S=S_0+S_2r^2$ , and  $\omega_p^2=\omega_{p0}^2+\omega_{p2}^2r^2$ , where  $S_0$  and  $S_2$  are the functions of z and  $\omega_{p0}^2=\omega_p^2(t,z,r=0)$ . The equations for evolution of plasma frequency  $\omega_p$  and beam width parameter  $f_1$  are

$$\frac{\partial(\omega_{p0}^{2} / \omega_{pm}^{2})}{\partial \eta'} = (1 - \omega_{p0}^{2} / \omega_{pm}^{2})G, \qquad (3)$$

$$\frac{\partial(\omega_{p2}^2 / \omega_{pm}^2)}{\partial \eta'} = -(\omega_{p2}^2 / \omega_{pm}^2)G - \frac{Gg_1}{2f_1^2}(1 - \omega_{p0}^2 / \omega_{pm}^2), \tag{4}$$

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$$\frac{\partial^2 f_1}{\partial \xi^2} = \frac{1}{f_1^3} - (\frac{\omega_{p2}^2}{\omega_{pm}^2}) (\frac{r_0^2 \omega_{pm}^2}{c^2}) f_1$$
 (5)

where  $G=\exp[(g-g_1)/\sqrt{f_1}]$ ,  $g_1=gf_1$ ,  $g=E_A/E_{00}$ . In Eqs. (3) and (4), G characterizes the tunnel ionization rate and fall off rapidly when beam width parameter  $f_1$  rises. At the front of the laser pulse  $(\eta=0)$ , the beam would diverge only due to diffraction.

The laser imparts an oscillatory velocity to electrons.

$$\vec{v}_1 = \frac{e\vec{E}_1}{mi\omega_1}.$$
 (6)

 $\vec{v}_1$  and  $\vec{B}_1$  beat to exert a ponderomotive force  $\vec{F}_1 = -(e/2c)\vec{v}_1 \times \vec{B}_1$ , on the electrons at  $2\omega_1$ , giving an oscillatory velocity  $\vec{v}_2$  at  $2\omega_1$ ,

$$\vec{v}_2 = \frac{\vec{F}_1}{mi \, 2\omega_1}.\tag{7}$$

It also produces density perturbation  $n_1$  at  $2\omega_1$ , in compliance with the equation of continuity. We get,

$$n_{1} = -\frac{n_{0}e^{2}k^{2}E_{1}^{2}}{4m^{2}\omega_{1}^{4}}$$
 (8)

Density perturbation  $n_1$  couples with  $\vec{v}_1$  to produce nonlinear current density at third harmonic  $\vec{J}_3^{NL} = -\frac{1}{2}n_1e\vec{v}_1$ .

$$\vec{J}_{3}^{NL} = \frac{n_{0}e^{4}k_{1}^{2}E_{1}^{3}}{8i\omega^{5}m^{3}}\hat{x}.$$
(9)

The nonlinear current produces third harmonic wave with electric field  $\vec{E}_3$ . This field also produces a self-consistent current  $\vec{J}_3^{\ L}$ ,

$$\vec{J}_{3}^{L} = -\frac{n_{0}e^{2}\vec{E}_{3}}{3\,m\,i\,\omega_{1}}.$$
(10)

The wave equation for the second harmonic field  $\vec{E}_3$  can be deduced from Maxwell's equations,

$$\nabla^2 \vec{E}_3 = \frac{4\pi}{c^2} \frac{\partial \vec{J}_3}{\partial t} + \frac{1}{c^2} \frac{\partial^2 \vec{E}_3}{\partial t^2},\tag{11}$$

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$$\nabla^2 \vec{E}_3 - \left[ \frac{9\,\omega_1^2 - \omega_p^2}{c^2} \right] \vec{E}_3 = -\frac{4\pi i 3\,\omega_1}{c^2} \vec{J}_3^{NL}. \tag{12}$$

Let the complementary solution of this equation is

$$E_{31} = A_3 \exp\{-ik_3S_3\} \exp[-i(3\omega_1 t - k_3 z)], \tag{13}$$

where  $S_3$  is a function of  $\emph{r}$  and  $\emph{z}$  . Using Eqs.(9) and (10), and equating real and imaginary parts

$$2\frac{\partial S_3}{\partial z} - \left(\frac{\partial S_3}{\partial z}\right)^2 - \left(\frac{\partial S_3}{\partial r}\right)^2 + \frac{1}{k_3^2 A_3} \left(\frac{\partial^2 A_3}{\partial r^2} + \frac{1}{r} \frac{\partial A_3}{\partial r}\right) - 1 + \frac{(9\omega_1^2 - \omega_p^2)}{k_3^2 c^2} = 0 \tag{14}$$

$$-\frac{\partial A_3^2}{\partial z}\frac{\partial S_3}{\partial z} + \frac{\partial A_3^2}{\partial z} - \frac{\partial A_3^2}{\partial r}\frac{\partial S_3}{\partial r} - A_3^2 \left(\frac{\partial^2 S_3}{\partial r^2} + \frac{1}{r}\frac{\partial S_3}{\partial r}\right) = 0$$
 (15)

In the paraxial ray approximation,  $r^2 << r_0^2 f_1^2$ , we expand  $S_3$  as  $S_3 = \phi(z) + \beta_3 r^2 / 2$ , where  $\beta_3^{-1}$  represents the radius of curvatures of the wave front of the third harmonic wave. For an initially Gaussian beam, we may write

$$A_3^2 = \frac{A_{30}^2}{f_2^2(z)} \exp[-3r^2 / r_0^2 f_3^2]$$
 (16)

Substituting the values of  $S_3$  and  $A_3^2$  in Eqs. (11) and (12) and equating the coefficients of  $r^2$  on both sides, we obtain

$$\beta_3 = \frac{1}{f_3} \frac{df_3}{dz},\tag{17}$$

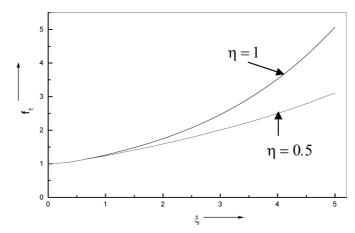
$$\frac{d^2 f_3}{dz^2} = \frac{1}{k_3^2 r_0^4 f_3^3} - \frac{\omega_{p2}^2 f_3}{c^2 k_3^2 r_0^2},\tag{18}$$

Now we suppose the solution (particular integral) of Eq.(12) is

$$E_{22} = A_2' \exp[-i(3\omega_1 t - 3k_1 z)], \tag{19}$$

where  $A_3' = A_{30}'(z)\psi_3$ ,  $\psi_3 = \exp[-r^2/r_0^2f_1^2]\exp[-3ik_1S_1]$ . Using Eq.(12) and (19), we obtain

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**Figure 1:** Variation of beam width parameter  $f_1$  as a function of normalized distance of propagation  $\xi$  for different values of  $\eta$ , the other parameters are:  $eA_{10}$  /  $m\omega_1 c = 0.02$ ,  $\omega_{pm} r_0$  /  $c \approx 20$  and  $k_1 c$  /  $\omega_1 = 0.44$ .

$$6ik_{1}\psi_{3}\frac{\partial A'_{30}}{\partial z} + \left[\frac{9\omega_{1}^{2} - \omega_{p}^{2}}{c^{2}} - 9k_{1}^{2}\right]A'_{30}\psi_{3} + A'_{30}\frac{\partial^{2}\psi_{3}}{\partial r^{2}} + A'_{30}\frac{1}{r}\frac{\partial\psi_{3}}{\partial r}$$

$$= -\frac{3\omega_{p}^{2}e^{2}k_{1}^{2}A_{10}^{3}}{8c^{2}\omega_{1}^{4}m^{2}f_{1}^{3}}\exp[-3r^{2}/2r_{0}^{2}f_{1}^{2}]. \tag{20}$$

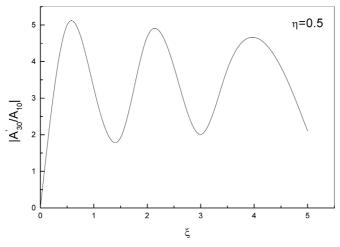
Multiply above equation by  $\psi_3^* r dr$  and integrate with respect to r, we get

$$\frac{\partial A_{30}''}{\partial \xi} - \left[ \frac{5}{3i} \left( \frac{\omega_{p0}^{2}}{\omega_{pm}^{2}} \right) \left( \frac{\omega_{pm}^{2} r_{0}^{2}}{c^{2}} \right) + \frac{1}{18i} \left( \frac{\omega_{pm}^{2} r_{0}^{2}}{c^{2}} \right) \left( \frac{\omega_{p2}^{2}}{\omega_{pm}^{2}} \right) f_{1}^{2} - \frac{1}{2i f_{1}^{2}} - \frac{1}{2i f_{1}^{2}} - \frac{1}{2i f_{1}^{2}} \left( \frac{\partial f_{1}}{\partial \xi} \right) - \frac{1}{2i} \left( \frac{\partial f_{1}}{\partial \xi} \right)^{2} \right] A_{30}'' = \left( \frac{eA_{10}}{mc\omega_{1}} \right)^{2} \left( \frac{\omega_{pm}^{2} r_{0}^{2}}{c^{2}} \right) \left( \frac{k_{1}^{2} c^{2}}{\omega_{1}^{2}} \right) \times \left[ \frac{1}{16i f_{1}^{3}} \left( \frac{\omega_{p0}^{2}}{\omega_{pm}^{2}} \right) + \frac{1}{48i f_{1}} \left( \frac{\omega_{p2}^{2}}{\omega_{pm}^{2}} \right) \right] \tag{21}$$

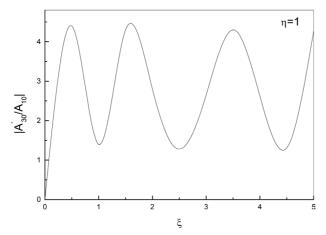
Where  $A_{30}''=A_{30}'/A_{10}$ , we have solved coupled Eqs. (3), (4), (5) and (21) numerically by applying the initial conditions; at  $\eta=0$ ,  $\omega_{p0}^2=0$ ,  $\omega_{p2}^2=0$ , for all  $\xi$ ; at  $\xi=0$ ,  $f_1=1$ ,  $\partial f_1/\partial \xi=0$  for all  $\eta$ . The parameters are  $E_A/E_{00}=3$  corresponding to  $I_0=21eV$  and laser intensity  $I=6\times10^{17}\,W/cm^2$ . We have plotted in Fig. 1,  $f_1$  Vs  $\xi$  or different values of  $\eta$ , the other parameters are  $eA_{10}/m\omega_1c=0.02$ ,  $\omega_{pm}r_0/c\approx20$  and

 $k_{_1}c$  /  $\omega_{_1}=0.44$ . One may note that at higher value of  $\eta$ , self-defocusing of main laser pulse increases. In Figs. (2) and (3), we have plotted normalized amplitude of the third harmonic wave  $A_{_{30}}''$  with normalized propagation distance  $\xi$ , for different values of  $\eta$ . It

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**Figure 2:** Variation of normalized third harmonic amplitude as a function of normalized distance of propagation  $\xi$  for  $\eta$ . The other parameters are:  $eA_{10}$  /  $m\omega_1c=0.02$ ,  $\omega_{pm}r_0$  /  $c\approx 20$  and  $k_1c$  /  $\omega_1=0.44$ .



**Figure 3:** Variation of normalized third harmonic amplitude as a function of normalized distance of propagation  $\xi$  for  $\eta=1$ . The other parameters are:  $eA_{10}$  /  $m\omega_1c=0.02$ ,  $\omega_{pm}r_0$  /  $c\approx20$  and  $k_1c$  /  $\omega_1=0.44$ .

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is noted that as the main laser pulse defocuses, the normalized third harmonic amplitude decreases with  $\xi$ , for stronger defocusing the third harmonic amplitude saturates.

### 3. DISCUSSION

Third harmonic generation of a Gaussian short pulse laser in a tunnel ionizing plasma is studied. An intense Gaussian laser beam propagating through a tunnel ionizing plasma gets self-defocused due to ionization induced refraction. A considerable decay in the intensity of third harmonic wave is obtained on account of self-defocusing of the fundamental wave. The third harmonic amplitude decrease initially and at the later time it saturates. The normalized third harmonic amplitude varies periodically with the distance with successive maxima acquiring lower value and saturate for stronger defocusing. The self-defocusing of the fundamental laser pulse decays the intensity of the third harmonic pulse.

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